IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of:

Group Art Unit: 1715

Torsten-Gottschalk-Gaudig et al.

Examiner:

Elena Tsoy Lightfoot

Serial No.:

10/738,543

Filed:

December 17, 2003

For:

WATER-WETTABLE SILYATED METAL OXIDES

Attorney Docket No.: WAS 0611 PUS

APPEAL BRIEF UNDER 37 C.F.R. § 41.37

Mail Stop Appeal Brief - Patents Commissioner for Patents U.S. Patent & Trademark Office P.O. Box 1450 Alexandria, VA 22313-1450

Sir:

This is an Appeal Brief from the final rejection of claims 15-19 and 30-37 of the Office Action mailed on May 17, 2010 for the above-identified patent application.

I. REAL PARTY IN INTEREST

The real party in interest is Wacker Chemie GmbH ("Assignee"), a corporation organized and existing under the laws of Germany, and having a place of business at Hanns-Seidel-Platz 4, München, Germany D-81737, as set forth in the assignment recorded in the U.S. Patent and Trademark Office on December 17, 2003 at Reel 014820/Frame 0734.

II. RELATED APPEALS AND INTERFERENCES

There are no appeals, interferences or judicial proceedings known to the Appellant, the Appellant's legal representative, or the Assignee which may be related to, directly affect or be directly affected by or have a bearing on the Board's decision in the pending appeal.

III. STATUS OF CLAIMS

Claims 15-19 and 30-37 are pending in this application. Claims 15-19 and 30-37 have been rejected and are the subject of this appeal. Claims 1-14 and 20-29 have been cancelled.

IV. STATUS OF AMENDMENTS

An amendment after final rejection was filed on August 24, 2010, and has been denied entry.

V. SUMMARY OF CLAIMED SUBJECT MATTER

Claim 15 claims partly hydrophobic silica particles (page 2, lines 4 - 5; page 3, line 5; page 5, lines 1 - 3) having a contact angle in air for water of less than 180°, a degree of coverage τ of the surface of the silica with silylating agent residues, based on the total silica particle surface area, of $1\% < \tau < 50\%$, a density of surface silanol groups SiOH ranging from 0.9 to 1.7 SiOH/nm² particle surface area, a carbon content of more than 0% and up to 2% by weight, and a methanol number less than 30 (page 12, lines 3 - 12), by silylating silica particles prepared under anhydrous conditions (page 3, line 9) with:

I) an organosilane of the formula

 $R^{1}_{n}SiX_{4-n}$

where n is 1, 2 or 3

or mixtures of these organosilanes,

- R¹ being a monovalent, optionally halogenated hydrocarbon radical having 1 to 24 carbon atoms, being identical or different at each occurrence, and being saturated, aromatic, monounsaturated, or polyunsaturated,
- X each independently being halogen, a nitrogen radical, OR^2 , $OCOR^2$, or $O(CH_2)_xOR^2$,
- R² being hydrogen or a monovalent hydrocarbon radical having 1 to 12 carbon atoms, and

```
x being 1, 2 or 3;
```

or

II) an organosiloxane composed of units of the formula

```
(R<sup>1</sup><sub>3</sub>SiO<sub>1/2</sub>), and/or
(R<sup>1</sup><sub>2</sub>SiO<sub>2/2</sub>), and/or
(R<sup>1</sup>SiO<sub>3/2</sub>)
```

where R¹ is as defined above,

the number of these units in one organosiloxane being at least 2; and I and II being used alone or in any desired mixtures in a total amount of from 0.015 mmol/g to 0.15 mmol/g per 100 m²/g of silica BET surface area.

(Claim 1 as filed, page 2, lines 1 to page 3, line 3).

The invention also relates, as claimed in claim 16, to a partly hydrophobic silica as claimed in claim 15, where the silylating is performed with the organosiloxane compound II (page 2, lines 17 - 23), and claim 18, where the silylation is perfumed with a silane (I) and an organosiloxane (II). (Page 2, lines 17 - 23).

The invention further pertains to partly hydrophobic silicas prepared by silylating with a silane of the formula (I) of claim 15, wherein, as in claim 19, the R¹ groups of the silane (I) are selected from methyl, ethyl, propyl, butyl, pentyl, hexyl, octyl, decyl, dodecyl, hexadecyl, octadecyl, phenyl, biphenyl, napthyl, benzyl, ethylphenyl, tolyl, and xylyl radicals. (Page 5, line 25 to page 6, line 14).

The invention further relates to partly hydrophobic silicas of claim 15, as claimed in claim 33, wherein the silylating agent is methyltrichlorosilane, dimethyldichlorosilane, trimethylchlorosilane, or hexamethyldisilazane (page 6, line 23 to page 7, line 9).

VI. GROUNDS OF REJECTION TO BE REVIEWED ON APPEAL

- A. Claims 15-17, 19, and 30-36 stand rejected for indefiniteness under 35 U.S.C. § 112 ¶2.
- B. Claims 15-19 and 30-37 stand rejected under 35 U.S.C. § 103(a) over Endo et al. Published application JP 04298538 A ("*Endo*"), in view of Fitzgerald et al. U.S. Patent No. 5,623,028 ("*Fitzgerald*") and Ward et al., U.S. Patent No. 5,578,189 ("*Ward*").

VII. ARGUMENT

The subject invention is directed to specific partly hydrophobic fumed silicas which have very surprisingly been found to be able to act as dispersants in the preparation of water-in-oil (W/O) and oil-in-water (O/W) dispersions, as well as for preparation of multiphasic dispersions such as oil-in-water-in oil (O/W/O) and water-in-oil-in-water (W/O/W) dispersions. See, e.g. page 18, lines 3 - 10 and Example 10 on page 25. The ability to form stable dispersions without the use of surfactants such as non-ionic polyoxyalkylene polyethers or anionic surfactants such as petroleum sulfonates or the like is particularly surprising, and highly useful, in particular in compositions where such conventional surfactants might prove disadvantageous. Examples are coatings, where the hydrophilic portion of conventional surfactants can render the coating hydrophilic and therefore less water resistant, and cosmetics, where many surfactants are also skin irritants to all or some.

Appellants discovered that silicas which meet the requirements of claim 15 are able to perform as a dispersing agent. The partly hydrophobic silicas are fumed silicas silylated with a silane (I), an organosiloxane (II), or a mixture of these, and the partly hydrophobic silica resulting from silylation must meet six different requirements:

- 1) a contact angle θ in air for water of less than 180°;
- 2) a degree of coverage τ of the silica surface, based on total surface area, of $1\% < \tau < 50\%$;

- a density of surface silanol groups of 0.9 to 1.7 SiOH/nm² (SiOH groups per square nanometer);
- 4) a carbon content greater than 0 and up to 2.0 weight percent;
- 5) a methanol number of less than 30; and
- 6) the silylating agents used to silylate the silica are used in amounts of 0.015 mmol/g to 0.15 mmol/g per 100 m²/g of silica BET surface area.

A. The Rejection Under 35 U.S.C. § 112 ¶2.

The Office contends that the language of claim 15 is indefinite. The language criticized by the Office has to do with the silylating agents I) and II) used in preparing the partly hydrophobic silica of the invention. In line 8 of claim 15, the claim recites that "at least one of I) and II)" are used in silylating the fumed silica. In lines 25 and 26 of the claim, it is recited "I) and II) being used alone or in any desired mixtures in a total amount of from 0.015 mmol/g to 0.15 mmol/g..." It is the position of this office, as Appellants understand it, that these two phases are in some manner indefinite. Appellants respectfully disagree.

The claims are addressed to one skilled in the art, and one so skilled has no problem with understanding the metes and bounds of the claim. Two types of silylating agents are possible for use in the claimed invention: silylating agent I) and silylating agent II). Line 8 indicates that at least one of these must be used, i.e. I) alone, II) alone, or a mixture of I) and II). Lines 25 and 26 specify the amounts which must be used. The total amount, which may be I) alone, II) alone, or I) and II) in any desired mixture, must total 0.015 - 0.15 mmol/g. The claim is clear, and is subject to but a single interpretation. It clearly meets the requirements of § 112 ¶2 to "particularly point out and distinctly claim." Reversal of the rejection of the claims under 35 U.S.C. § 112 ¶2 is respectfully solicited.

B. Rejection of the Claims Under 35 U.S.C. § 103(a)

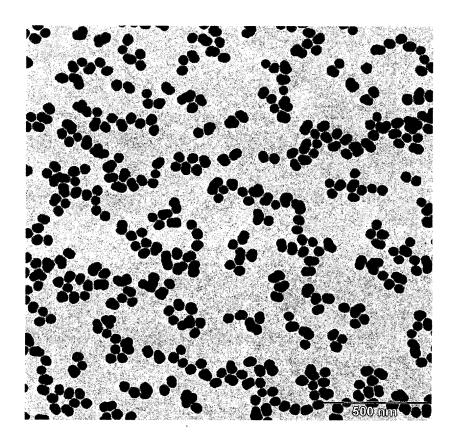
All the claims have been rejected under 35 U.S.C. § 103(a) over *Endo* in view of *Fitzgerald* and *Ward*.

Endo is directed to preparing polyester films with improved running characteristics while maintaining transparency. The Office has provided an English language translation of the underlying Japanese language document. The translation does not appear to be a machine translation, but portions are difficult to understand, as they appear to be in sharp conflict with well known properties of silica.

The "running" characteristics referred to by *Endo* have to do with processing of polyester films, which are processed by means of rollers, both in stretched (oriented) and unstretched (non-oriented) films. The films may also be biaxially stretched. To improve running characteristics, *Endo* states that microparticles (fillers) have been used in the past to roughen the surface, but have not proven to be satisfactory. ¶[0004]. Among these fillers are polyester catalyst degeneration residues and fillers such as "calcium carbonate, titanium dioxide calcium phosphate, or other inorganic compound particles." ¶[0005]. In ¶[0006], the use of "silica particles that have relatively good affinity with polyester", i.e. hydrophobicized silicas, are cited as having been used by the prior art, but subject to drawbacks. Some of the problems associated with silica fillers are poor dispersibility, decrease in transparency, and a decrease in the polyester polymerization rate during manufacture of the polyester ¶[0006].

Thus, *Endo* sought to improve running characteristics while avoiding the aforesaid drawbacks by the use of a special solution-processed silica ("colloidal silica") which has silane coupling agents bearing reactive organic functional groups grafted to the surface of the silica. As is well known, colloidal silica consists of spherical particles, and ordinarily has particle sizes in the low nm range. Such colloidal silicas are widely used as the abrasive in chemical mechanical polishing of silicon wafers, for example. However, the particles of *Endo* are required to be rather unconventionally larger, with a minimum particle size of 0.1 μ m (100 nm), and a maximum particle size of 5 μ m (5000nm) (¶[0008]), more preferably 0.5 μ m to 2 μ m (500 nm – 2 μ m (2000nm)) ¶[0016].

Endo indicates that the particles he uses are unlike other particles, which can be destroyed by stretching during film orientation. ¶[0005]. This is not surprising, since colloidal silica particles are spherical and uniform, as is well known. A photomicrograph of colloidal silica is shown below. The uniformity and high sphericity are notable.



Reference may also be made to the "colloidal silica bible", R.K. Iler, The Colloid Chemistry of Silica and Silicates, Cornel University Press, Ithaca, New York, 1955. This reference is now some five decades old, and its teachings are so well known that judicial notice may be taken thereof.

Spherical particles have no anisotropy. Therefore, they react to stress in the same manner, regardless of the direction from which the stress is applied. This is one reason that the specific particles of *Endo* are not destroyed during stretching. Non-spherical

particles, especially elongate particles, can easily be destroyed in stretching. Spherical particles are also non-orientable: They cannot be oriented during sketching of the film due to the fact that they are spherical.

However, *Endo* indicates that the use of spherical, colloidal silica particles of large size, alone, is not sufficient. The particles must also be functionalized to both reduce the surface silanol content as well as providing an organic functional group ("coupling group") reactive with the polyester, by reacting the colloidal silica with a compound of the formula YRSiX₃ where Y is a reactive functional group such as vinyl, methacryl, epoxy, or amino, although other functional groups may be used, linked to silicon by R, an alkyl group (actually an alkylene group), and X is a hydrolyzable group such as an alkoxy group. The functional groups react, or "couple" the functionalized colloidal silica to the polyester matrix. ¶[0013].

At the end of $\P[0012]$, *Endo* indicates that silica produced by "the gas phase method", i.e. fumed silica, is <u>not</u> acceptable, since the polymerization will be delayed, a problem discussed with other fillers in $\P[0006]$.

The disclosure of *Endo* is not well understood, and has numerous inconsistencies which are at sharp contrast to what is well known to those skilled in the art.

For example, *Endo* discloses that his colloidal silica particles have a surface silanol content of 1-30/nm² (1-30 SiOH per square nanometer). However, colloidal silica is prepared in an aqueous environment and conventionally has a surface silanol content of 4-5 SiOH/nm². See, e.g. A.P. LeGrand, The Surface Properties of Silicas, John Wile & Sons, Chichester, 1998. The fumed silicas exemplified by *Endo* have surface silanol contents above this conventional range, 7.0 and 8.0 SiOH/nm². This corresponds to the maximum silanol content as calculated and measured by numerous researchers, as reported in Iler, *Op. Cit.*, pages 241-247, where the maximum, assuming 100% surface courage by silanol groups is 8. However, colloidal silicas having SiOH group contents appreciably lower than 4 are unknown, and *Endo* does not disclose how any colloidal silicas having this low silanol

content could be prepared.¹ Endo does not enable production of silicas with the low silanol contents he describes. Moreover, Endo indicates [0015] that if the silanol content of the starting silica is below 1 SiOH/nm², the particles have no dispersibility in water or alcohols, and yet it has been shown by applicants that silica having such an SiOH content is a least partly dispersible in water, and fully dispersible in methanol, for example. Endo also discloses that if the silanol content is greater than 30 SiOH/nm², dispersibility in water and alcohol is poor, and yet it is well known that the higher the number of surface silanol groups, the higher the polarity, and the higher the dispersibility. These inconsistencies of Endo would lead one skilled in the art to question the technical validity of the entire Endo disclosure.

Endo does not disclose the use of <u>fumed</u> silica² treated with a coupling agent, and the office has supplied *Fitzgerald* to allegedly supply this deficiency. The Office cites *Fitzgerald* for disclosing a "raw" (untreated) fumed silica having a BET surface area of 200 m²/g and a surface silanol content of 4.5 SiOH/nm², which may be treated with silane to reduce the surface silanol content, to use as a filler in "resin compositions" (Office Action of May 17, 2010, page 4). Applicants are not sure where the term "resin compositions" comes from, as *Fitzgerald* is not directed to silicone resins, a term of art quite distinct from the use of the term "resin" in other fields³, but is directed to silicone rubber formulations.

Ward is cited for the proposition that 200 m²/g surface area commercial silica has a particle size of 0.20-0.35 microns. This is partially correct. Ward is directed to the use of fumed silica of reduced particle size in injection moldable polymers. The starting silica has a surface area of from 50 to 600 m²/g and an initial aggregate size (not particle size) of 0.10 to 0.25 μ m (col. 2, line 56). However, Ward teaches that such silica is unsuitable for

¹ How colloidal silicas can be prepared with more than 8 SiOH/nm² is also mystifying.

² Fumed silica, as is well known, generally has about 2.5 to 3 SiOH/nm² as prepared. These values may range higher if the silica is not prepared under anhydrous conditions or is allowed to adsorb water following preparation.

³ As is well known in the field of silicons, silicone resins are non-linear, highly crosslinked, three dimensional polymers containing T and Q units and generally M units as well. T resins are also called silsesquioxane resins. T resins, MT resins, MQ resins and MTQ resins are all common silicone resins. None of these are linear "resins" as that term is used for other polymers such as polypropylene and polyesters, all of which are linear polymers.

injection molding of polymers, and teaches that the aggregates should be broken down under high shear to smaller particle sizes. No ultimate aggregate sizes are given, but it is clear that Ward teaches away from using particles⁴ of 0.1 μ m or larger, since the minimum size of the starting aggregates is 0.1 μ m, and Ward teaches high shear size reduction of these to smaller sizes.

The Office states:

Therefore, it would have been obvious to one of ordinary skill in the art at the time the invention was made to have used typical commercially available raw fumed silica having particle size of 0.20-0.35 microns as raw silica in *Endo* et al for treating with silane coupling agent with the expectation of producing silica having excellent dispersibility in a polyester film composition since *Endo* et al does not limit its teaching to particular raw silica, and commercially available raw fumed silica has particle size, a surface area and a surface silanol density within ranges required in raw silica of *Endo* et al.

However, *Endo* is very specific about his silica, which is a colloidal silica of relatively large particle size, and indicates that prior attempts to use other fillers, including other silicas, even silicas compatible with the polyester matrix, are deficient. *Endo* thus teaches away from using other silicas, e.g., silicas other than large size, colloidal silica of spherical morphology. Teaching away is strong evidence of non-obviousness, *W.L. Gore v. Garlock*, 721 F.2d 1540 (Fed Cir 1983). A further teaching away is found in ¶[0012] wherein silica produced by the gas phase method, i.e. fumed silica, is said by *Endo* to be unsuitable.

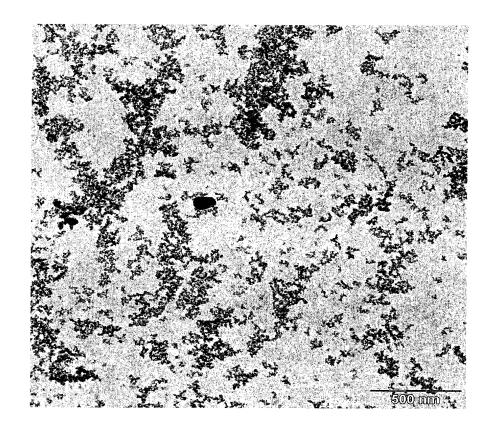
⁴ Three different "particles" are involved with reference to fumed silica. Modernly, these are known as primary particles; aggregates, or "secondary particles"; and agglomerates. Primary particles essentially do not exist in fumed silica. They are formed in the high temperature flame, but immediately sinter or fuse into aggregates, or "sinter aggregates". These particles are difficult to reduce in size since they are quite small, and consist predominately of primary particles fused together in amorphous, irregular structures. Upon collection and further processing, these aggregates collect into agglomerates. Agglomerates can easily be broken down into aggregates under high shear. It is believed, for these reasons, that the term "aggregate" as used by ward is actually "agglomerate."

The subject invention is not directed to the problem addressed by *Endo*, improving the running characteristics of polyester films, but is directed to the problem of providing aqueous dispersions which are stable without the use of conventional emulsifiers and surfactants. This was surprisingly and unexpectedly achieved by the use of partly waterwettable furned silicas, a problem totally remote from that of *Endo*.

Appellants respectfully submit that there is no prima facie case of obviousness here. It is well settled that teachings of a reference can only be modified when there is some objective teaching or suggestion in the references themselves or knowledge generally available to one of ordinary skill in the art as to the desirability or incentive of such a modification. See, e.g. MPEP § 2143. If the prior art provides no reason for one of ordinary skill in the art to modify the prior art as claimed, the modification would not have been obvious.

Endo is directed to improving running properties of polyester film, and does so by using a specific, large size, spherical colloidal silica, reacted with a hydrolysable silane to provide organic functional groups which couple with the polyester. Endo states that other fillers, including other silicas, even treated silicas, do not work. Endo does not teach or suggest the use of other silicas, but rather teaches away from their use.

Fitzgerald is not directed to improving the running characteristics of polyester film as is Endo, but is directed to the use of pyrogenic silica (fumed silica) to improve the compression set of silicone elastomers. Where is the motivation to combine these references? Fumed silica has an irregular, chain like structure rather than the isotropic, spherical morphology of colloidal silica. This structure of fumed silica is well known, and photomicrographs are widely available, both from the manufacturers as well as literature sources, the internet, etc. A photomicrograph of typical fumed silica is reproduced below.



The elongate structure of fumed silica could be broken down by the stretching of the polyester film of *Endo*, a phenomenon *Endo* teaches avoiding. There is no motivation to combine the compression set-improving properties of the hydrophobicized fumed silica of *Fitzgerald* with the running characteristic-improving, coupling agent-treated colloidal silica of *Endo*.

The same is true of *Ward*. Ward indicates that the <u>starting</u> "aggregate" size of fumed silica can be as low as $0.1 \mu m$, and teaches reducing this size to lower values, values which, since they are lower than $0.1 \mu m$, are lower than the lowest range of colloidal silica stated to be useful by *Endo*. See ¶[0016] of *Endo*.

Thus, it is respectfully submitted that the references teach away from their combination, and no prima facie case of obviousness has been established.

The claimed particles contain five physical property limitations. These are

- 1) a content angle θ in air for water of less than 180°;
- 2) a degree of coverage τ of 1% 50%;
- a density of surface silanol groups between 0.9 and 1.7 SiOH/nm²;
- 4) a carbon content of greater than 0% and up to 2.0% by weight; and
- 5) a methanol number of less than 30.

In addition, the claim requires the silica to have been silylated with silanes I), II), or a mixture thereof.

The combination of *Endo*, *Fitzgerald*, and *Ward*, even were this combination proper, do not disclose, teach, or suggest these limitations. <u>None</u> of the references even mentions contact angle, carbon content, or methanol number. *Endo* mentions degree of coverage, but for colloidal silica, not for fumed silica.

Fitzgerald mentions density of surface silanol groups in silylated fumed silica, but the lowest value reported is 2.39. Fitzgerald does not disclose any value even close to Appellants maximum value of 1.7, nor does he enable the preparation of such a silica.

Nor do the references inherently disclose these limitations, and even if they did, such an inherent disclosure is irrelevant, as inherency applies only to rejections for anticipation under 35 USC § 102, not for obviousness under 35 USC § 103. See, e.g. *In re Spormann*, 363 F.2d 444 (CCPA 1966); *In re Naylor*, 369 F.2d 765 (CCPA 1966); and *In re Shetty*, 566 F.2d 81 (CCPA 1977). Rather, there must be an actual disclosure, teaching, or suggestion. That is not the case here.

Moreover, even if inherency did apply, the inherency must be certain, and must be the necessary and <u>only</u> result, not merely a possible result or even a probable result. In re Robertson, 169 F.3d 743 (Fed. Cir. 1999). There is no such inherency here. The examples of *Endo* bear this out. First they are directed to colloidal silica, and not fumed silica, and even were this distinction ignored, contrary to the teachings of *Endo*, no example of *Endo* falls within appellants' claimed range of SiOH content. The lowest number of silanol groups in the inventive Examples of *Endo* is in Example 3, where 65% of the 8.0

SiOH/nm² were reached, leaving 2.45 SiOH/nm². This is well above applicants claimed range.

In Comparative Example 3, *Endo* reacts 85% of surface hydroxyl groups of a colloidal silica having a surface silanol content of 7 SiOH/nm² and otherwise having all the characteristics he discloses as useful, but this high amount of coupling agent derivatization, leaving only 1.2 SiOH/nm², is stated to be unsuitable for use in his invention.

Claim 16 requires the silylating agent to be an organosiloxane compound (II). *Endo* does not teach or suggest the use of such a silylating agent. All the silylating agents taught by *Endo* are hydrolyzable silanes, not organosiloxanes. Claim 18 requires silylating with both a silane and an organosiloxane. *Endo* does not teach any silica silylated with both these silylating agents. Claims 16 and 18 are separately patentable.

Claim 19 requires silylating with a silane in which R¹ is methyl, ethyl, propyl, butyl, pentyl, hexyl, octyl, decyl, dodecyl, hexadecyl, octadecyl, phenyl, biphenyl, napthyl, benzyl, ethylpenyl, tolyl, or xylyl. *Endo* requires a functional organic group so that the particles can be coupled to the polyester. None of the R¹ groups of claim 19 are functional organic groups. The same is true of claim 33, wherein specific silylating agents are used. Even if the use of the specific type of colloidal silica required by *Endo* were disregarded, *Endo* still requires an organic functional group with which to couple the silica to the polyester. One skilled in the art would not be motivated to silylate silica with a nonfunctional silane for use in the compositions of *Endo*. Claims 19 and 33 are separately patentable.

In summary, there is no motivation to combine *Endo*, *Fitzgerald*, and *Ward*, and *prima facie* obviousness has not been established. *Endo* teaches away from the proposed combination. The claims are directed to parly water wettable fumed silica particles which are not only silylated by specific silylating agents I) and II), but also are characterized by five physical property limitations, at least three of which are not even mentioned by the references.

The fee of \$540.00 as applicable under the provisions of 37 C.F.R. § 41.20(b)(2) is <u>not</u> enclosed per §1204.01 Reinstatement of Appeal [R-3] ruling. Appeal Fees were filed in earlier prosecution of this case and should be applied to this new appeal. Please charge any additional fee or credit any overpayment in connection with this filing to our Deposit Account No. 02-3978.

Reversal of all rejections of record is respectfully requested.

Respectfully submitted,

TORSTEN-GOTTSCHALK-GAUDIG ET AL.

William G. Conger

Reg. No. 31,209

Attorney/Agent for Applicant

Date: November 12, 2010

BROOKS KUSHMAN P.C. 1000 Town Center, 22nd Floor Southfield, MI 48075-1238

Phone: 248-358-4400 Fax: 248-358-3351

Enclosure - Appendices

VIII. <u>CLAIMS APPENDIX</u>

Claims 1 - 14. (Cancelled).

15. Partly hydrophobic fumed silica particles, said partly hydrophobic fumed silica particles having a contact angle θ in air for water of less than 180°, a degree of coverage τ of the surface of the silica with silylating agent residues, based on the total silica particle surface area, of $1\% < \tau < 50\%$, a density of surface silanol groups SiOH ranging between a minimum of 0.9 and a maximum of 1.7 SiOH/nm² particle surface area, and having a carbon content of more than 0% and up to 2.0% by weight, and a methanol number of less than 30, said partly hydrophobic silica prepared by a process comprising silylating fumed silica particles prepared under anhydrous conditions, with at least one of I) and II)

I) an organosilane of the formula

 $R^{1}_{n}SiX_{4-n}$

where n is 1, 2 or 3

or mixtures of these organosilanes,

- R¹ being a monovalent, optionally halogenated hydrocarbon radical having 1 to 24 carbon atoms, being identical or different at each occurrence, and being saturated, aromatic, monounsaturated, or polyunsaturated,
- X each independently being halogen, a nitrogen radical, OR^2 , $OCOR^2$, or $O(CH_2)_xOR^2$,
- R² being hydrogen or a monovalent hydrocarbon radical having 1 to 12 carbon atoms, and
- x being 1, 2 or 3;
 - II) an organosiloxane composed of units of the formula

 $(R^1_3SiO_{1/2})$, and/or

Appendix

 $(R^1_2SiO_{2/2})$, and/or

 $(R^1SiO_{3/2})$

where R¹ is as defined above, or mixtures thereof,

the number of these units in one organosiloxane being at least 2; and I and II being used alone or in any desired mixtures in a total amount of from 0.015 mmol/g to 0.15 mmol/g per 100 m²/g of silica BET surface area measured by the BET method in accordance with DIN 66131 and 66132.

16. The particles of claim 15, wherein said silylating is performed with an organosiloxane composed of units of the formula (II)

 $(R^1_3SiO_{1/2})$, and/or

(R¹₂SiO_{2/2}), and/or

 $(R^1SiO_{3/2})$

where R¹ is as defined above, or mixtures thereof,

the number of these units in one organosiloxane being at least 2; II being used in a total amount of from 0.015 mmol/g to 0.15 mmol/g per 100 m²/g of silica BET surface area measured by the BET method in accordance with DIN 66131 and 66132.

17. The particles of claim 15, wherein said silylating is performed with an organosilane of the formula

 $R^{1}_{n}SiX_{4-n}$

where n is 1, 2, or 3, or a mixture of these organosilanes, where R^1 is a $C_{1\cdot 24}$ hydrocarbon radical selected from the group consisting of alkyl radicals, alkenyl radicals, aryl radicals, and alkylaryl radicals, each R^1 being the same or different,

- X each independently being halogen, a nitrogen radical, OR^2 , $OCOR^2$, or $O(CH_2)_xOR^2$,
- R² being hydrogen or a monovalent hydrocarbon radical having 1 to 12 carbon atoms, and

x being 1, 2 or 3.

- 18. The particles of claim 15, wherein said step of silylating is performed with a mixture of at least one organosilane of the formula $R^1_n SiX_{4-n}$ with an organosiloxane of the formula (II).
- 19. The particles of claim 17, wherein each R¹ individually is selected from the group consisting of methyl, ethyl, propyl, butyl, pentyl, hexyl, octyl, decyl, dodecyl, hexadecyl, octadecyl, phenyl, biphenyl, napthyl, benzyl, ethylpenyl, tolyl, and xylyl radicals.

20. - 29. (Cancelled).

- 30. The particles of claim 15, wherein said partly hydrophobic silica has a methanol number less than 20.
- 31. The particles of claim 15, wherein said partly hydrophobic silica has a carbon content of 0.1 to 0.5 weight percent per each 100 m²/g of surface area.
- 32. The composition of claim 17, wherein R¹ is independently selected from the group consisting of methyl, octyl, and vinyl.
- 33. The partly hydrophobic silica particles of claim 17, wherein at least one organosilane is selected from the group consisting of methyltrichlorosilane, dimethyldichlorosilane, trimethylchlorosilane, and hexamethyldisilazane.
- 34. The partly hydrophobic silica particles of claim 15, wherein the contact angle θ is between 100° and 0°.
- 35. The partly hydrophobic silica particles of claim 15, wherein the contact angle θ is between 90° and 0°.

- 36. The partly hydrophobic silica particles of claim 15, wherein the density of surface silanol groups is between 1.2 and 1.7 SiOH per nm² of particle surface.
- 37. The partly hydrophobic silica of claim 15, which is effective to stabilize water-in-oil and oil-in-water emulsions without also using an emulsifier.

IX. EVIDENCE APPENDIX

None

X. RELATED PROCEEDINGS APPENDIX

None